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Modelling the tropospheric and stratospheric sulfur isotopes in a column model for volcanically quiescent periods

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It is debated how much stratospheric sulfate aerosol (SSA) in volcanically quiescent times is replenished by carbonyl sulfide (COS) oxidation products. The atmospheric COS budget is also currently uncertain, with missing sources and sinks. Isotopic analysis can be used to allocate the missing sources of COS and also to further constrain the relevance of COS to SSA. The measured tropospheric isotopic signature of COS ($\delta^{34}S$) ranges from 10-14 ‰ (Kamezaki et al., 2019; Angert et al.,2019; Hattori et al., 2020; Davidson et al., 2020), whereas SSA $\delta^{34}S$ is constrained by only one single measurement at 18 km of 2.6 ‰ (Castleman, 1974). We use an atmospheric column model to constrain the COS isotopic budget and understand the contribution of COS to sulfate. We find that the COS tropospheric signal is determined by the signatures of its precursors (carbon disulfide, CS_2, and dimethyl sulfide, DMS) and fractionation during plant uptake and oxidation. Photolysis of COS is important in the stratosphere; the isotopic signal of COS propagates through sulfur dioxide (SO_2) to sulfate in the stratosphere. The model can reproduce $\delta^{34}S$ between 1-5 ‰ in the lower stratosphere, which encapsulates the observations from Castleman (1974).

References

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